

INFLUENCE OF POLYMER GRADE AND COMPATIBILIZER  
CONCENTRATION ON THE INTERFACIAL SHEAR STRENGTH  
IN SISAL FIBRE POLYPROPYLENE COMPOSITES

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ABSTRACT

In this work we investigated the interfacial shear strength for the system sisal polypropylene, regarding the presence of a compatibilizer (maleic anhydride grafted polypropylene) as well as the grade of polypropylene, namely two homopolymers with different flow behaviour. Composites containing different concentrations of sisal fibre and compatibilizer were produced using a co-rotating twin screw extruder and subsequently injection moulded to retrieve test specimens. These specimens were investigated for tensile and notched impact properties, as well as the final fibre length in the composites was determined. With this data, a simple rule of mixtures model was applied to evaluate the tensile strength and notched impact strength for the interfacial shear strength. Further, single fibre pull out tests have been carried out with sisal fibres from polypropylene.

Data for interfacial shear strength, acquired by the single fibre pull out tests fit the data acquired by the evaluation of the mechanical testing very well. Furthermore the presence of a compatibilizer improves the interfacial shear strength, as well as the polypropylene grade shows an influence, which is also found from the single fibre pull out tests. In conclusion this shows good applicability of single fibre pull out tests for estimating composite properties. However, one has to keep in mind several influences, due fibre orientation and processing method, when comparing the results from macromechanical measurements with single fibre pull out tests.

1. INTRODUCTION

Natural fibres in common are known as reinforcements in polymers for a long time. While the first intention to use materials like hemp, flax or wood in thermoplastic polymers was mainly related to their relatively low price, further developments due to their good mechanical performance in combination with their low density and renewability took place. Scientific and industrial interest in these materials increased steadily over the last years. Especially the role of the interface between fibre and matrix, which is known to be a dominant factor in glass fibre

reinforced composites (Thomason 2002, Thomason and Schoolenberg 1994), was investigated several times in literature. A method of characterization, which is often used, is the preparation of fibres with a certain surface treatment and the subsequent processing to yield a composite, which is tested for its properties. This method has the advantage that it shows the final properties in correlation to fibre treatment or additive dosage, but information about the interface, especially about the interfacial shear strength, is hard to acquire most times.

On the other hand, acquisition of the interfacial shear strength is possible via different micromechanical tests, like single fibre pull out or microfragmentation tests (Kim and Mai 1998). Such methods characterize the interface very well, (although there can be some serious problems in sample preparation, especially with thermoplastic matrices) but suffer from the problem that the estimation of composite properties from this key figure is very limited.

The aim of this work was to compare the interfacial shear strength acquired from single fibre pull out tests with the one calculated from the mechanical properties of the composite, especially regarding the influence of the presence of a compatibilizer in two different polypropylene grades.

## 2. MATERIALS AND METHODS

For this study, 10 – 50 wt% of sisal fibres (with cut length 6 mm) and 0 – 4 wt% compatibilizer (Exxelor PO1020, Exxon Mobile Chemicals) were compounded into the polypropylene matrix (Daplen HD120MO, MFR 8 g/10 min, and HC205TF, MFR 4 g/10 min, Borealis) by means of a co rotating parallel twin screw extruder Thermo Prism TSE24HC with a processing length of 28 D. The compounder was equipped with a gravimetrical dosing system. Throughput was 8 kg h<sup>-1</sup> and the barrel temperature was set to 200°C. All process parameters were held constant throughout the whole processing. The melt strands were cooled via a water bath and cut to granules. These granules were dried for at least 4 h in a circulating air oven at 80°C prior to injection moulding.

Injection moulding of universal test specimens was carried out on an Engel ES80 injection moulding machine in compliance with ISO 3167. Barrel temperature was set to 210°C. The specimens were stored at least for 88 h at 23°C and 50% relative humidity prior to mechanical testing. Specimens for impact testing were produced from the universal test specimen using a pneumatic punch.

Tensile strength and modulus were measured using a Zwick-Roell Z020 universal testing machine complying with ISO 527. Crosshead speed was set to 1 mm min<sup>-1</sup> for the acquisition of the elastic modulus, after that the speed was set to 2 mm min<sup>-1</sup> until the break of the samples. The impact specimens were notched with a Coesfeld moulding cutter and were tested on a Zwick/Roell 5113.300 impact testing machine according to ISO 179eA.

To regain the fibres from the polymer, 1 g of the sample, taken from the parallel part of the universal test specimen, was extracted in boiling xylene under reflux for 3 h (Takase and Shiraishi 1989). After that the hot solution was vacuum filtered to yield the fibres, which were dried afterwards. Fibre distribution scans were carried out on an Olympus BX61 light microscope with a CCD camera and a motorized, fully automated stage. The sisal fibres were illuminated by transmitted light. Image analysis was carried out with analySIS Five from Soft Imaging Systems to yield the length of the different fibres. To get reliable values, at least 500 fibres were detected with the image analysis. This was accomplished by multiple image alignment, a method where the microscope is taking multiple pictures by altering stage position

along a grid and these are assembled to get a bigger display window. The length weighted lengths were calculated from these results to get reliable key values for fibre length.

Samples for single fibre pull out tests were prepared from the raw sisal fibres with a cut length of 60 mm. The fibres were pushed into a molten polypropylene film with a thickness of about 1 mm. This embedding was done between two microscopic slides at a hot press. The polypropylene grades used were the same as for composite production. After the embedding, the samples were cooled down to room temperature. These samples were tested in the single fibre pull out test apparatus, which was built up from a Physikinstrumente stage, type M-403.DG, a Tensiometric load cell, type Stak 1390, with a range from 0 – 30 N, and a self assembled frame of aluminium profiles. This setup was designed to fit under the light microscope mentioned above, to measure the embedded length and the fibre diameter, as well as to get the possibility to monitor the whole pull out procedure. Furthermore this setup provides the possibility to inspect the quality of the interface, e.g. if voids or cracks are present between fibre and matrix, prior to measurement, to discard samples with poor quality. The measurements have been carried out at a crosshead speed of 1 mm min<sup>-1</sup>.

### 3. RESULTS AND DISCUSSION

The tensile strength of the different produced composites was evaluated using equation (1), a modified version of the Kelly-Tyson model (Kelly and Tyson 1964), valid only for subcritical fibres.

$$\sigma_c = \eta_o \tau_c \frac{l}{d} V_f + \sigma_m (1 - V_f), \quad (1)$$

where  $\sigma_c$  and  $\sigma_m$  are the composite and matrix strength,  $\eta_o$  is an orientation factor,  $\tau_c$  is the interfacial shear strength,  $V_f$  is the fibre volume fraction and  $l$  and  $d$  are fibre length and diameter. The orientation factor  $\eta_o$  was determined to be around 0.77 for injection moulded sisal fibre composites, as shown in a previous work (Burgstaller 2007). With this model, the results from mechanical testing and the fibre scans, it was possible to calculate the interfacial shear strength. Looking at the results from this calculation (Table 1), one can see that the interfacial shear strength for the composites containing some compatibilizer is increasing with increasing fibre content. Therefore we calculated the concentration of the compatibilizer in the matrix ( $C_m$ ), because we found good correlations between this concentration and the improvement in mechanical properties for wood plastic composites in a previous work (Burgstaller and Stadlbauer 2006).

The evaluation of the notched impact strength was done with the model shown in equation (2). This approach is based on the Cottrell model (Thomason and Vlug 1997), but again only accounts for subcritical fibres and the failure mode of fibre pull out.

$$a_{cNe,c} = \eta_o \tau_c \frac{l^2}{d} V_f + a_{cNe,m} (1 - V_f), \quad (2)$$

where  $a_{cNe,c}$  and  $a_{cNe,m}$  are the notched impact strength of the composite and the matrix, respectively,  $\tau_c$  is the interfacial shear strength,  $V_f$  the fibre volume fraction and  $l$  and  $d$  are fibre length and diameter. Applying this model to the results of the mechanical testing, we again find the interfacial shear strength (Table 1), which is in the same range as in the according calculation for tensile strength in this case.

An exemplary plot for a single fibre pull out of sisal from polypropylene HD120MO is shown in Figure 1. In this plot, the maximum force is 2.61 N, and with a fibre diameter of 0.239 and an embedded length of 1.036 one yields an interfacial shear strength of 3.36 MPa using  $\tau_c = F_{Max}/(\pi ld)$ .

To get reliable values, 20 samples have been subjected to this testing. These were inspected for voids and imperfections under the microscope prior to testing, and samples with such failures were discarded. The arithmetic mean for  $\tau_c$  from this set of samples is 6.21 MPa, with a standard deviation of 2.3 MPa.

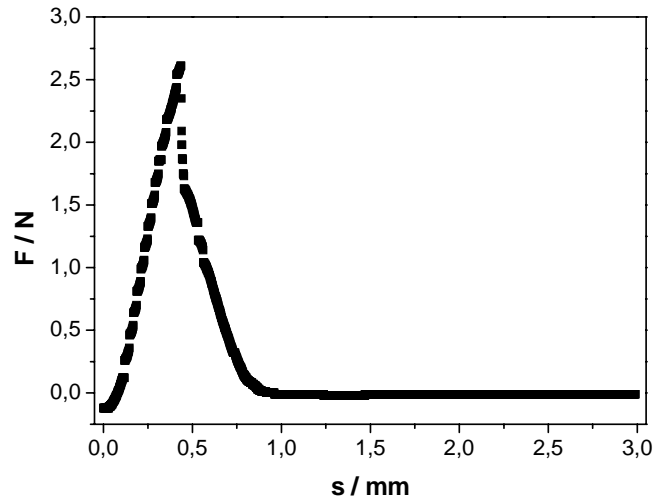


Fig. 1. Force – displacement curve for a pull out experiment with a single sisal fibre from polypropylene HD120MO

Table 1. Data from mechanical testing and interfacial shear strength calculated thereof

matrix	type	$C_c^{***}$	$C_m^{***}$	$C_{fibre}$	$V_f$	$l/d$	$\sigma_c$	$\tau_c^*$	$a_{CNe.c}$	$\tau_c^{**}$
[wt%]		[wt%]	[wt%]	[wt%]			[MPa]	[MPa]	[kJm <sup>-2</sup> ]	[MPa]
100				0	0	--	32.3	--	2.04	--
90				10	0.070	10.4	32.3	2.69	4.18	2.54
80		0	0	20	0.145	10.5	31.6	4.48	6.18	3.04
70				30	0.266	11.1	32.9	5.38	8.59	3.57
60				40	0.325	11.2	36.0	5.21	8.06	5.12
50				50	0.445	11.0	36.5	5.18	7.35	4.51
88			2.04	10	0.062	8.7	33.7	7.86	2.73	8.64
78			2.27	20	0.130	10.8	35.1	6.30	3.87	6.94
68		2	2.56	30	0.246	12.1	36.9	5.40	5.39	5.93
58			2.94	40	0.373	9.9	44.4	8.47	5.98	9.32
48			3.45	50	0.423	10.8	48.4	8.48	5.34	9.32
86		4	4.17	10	0.078	10.3	32.2	6.27	3.01	6.90
76			4.65	20	0.174	10.1	37.7	9.12	4.27	10.04

## Influence of polymer and compatibilizer on interfacial shear strength

66		5.26	30	0.266	10.3	45.4	10.83	5.10	11.91
56		6.06	40	0.349	10.2	50.3	11.06	5.40	12.16
46		7.14	50	0.443	10.0	54.8	11.07	5.31	12.17
100			0	0	--	37.3	--	1.80	--
90			10	0.078	8.3	36.1	3.5	3.79	3.83
80	0	0	20	0.162	10.3	36.5	4.1	5.55	4.49
70			30	0.268	10.4	36.4	4.2	6.94	4.64
60			40	0.367	10.1	36.5	4.5	7.55	4.97
50			50	0.489	9.6	33.8	4.1	6.47	4.51
88		2.04	10	0.082	10.1	38.3	6.5	13.54	7.16
78		2.27	20	0.135	10.8	42.6	9.3	12.89	10.26
68	2	2.56	30	0.295	9.8	47.8	9.7	10.13	10.65
58		2.94	40	0.391	10.3	50.6	9.0	11.18	9.92
48		3.45	50	0.496	10.3	62.2	11.1	5.41	12.16

\* calculated from tensile strength, \*\* calculated from impact strength, \*\*\*  $C_c$  is the concentration of the compatibilizer in the whole formulation,  $C_m$  is the concentration in the matrix

As can be seen in Table 2, the two different polymers yield different interfacial shear strengths. The polypropylene HD120MO, which is the less viscous grade of the two, shows higher average interfacial shear strength than the more viscous grade. This should occur due to better wetting of the fibres with the less viscous grade. Interesting to note is that this trend is also observed from the evaluation of the tensile and impact strength, where the more viscous grade yields  $4.08 \pm 0.36$  MPa interfacial shear strength versus  $5.06 \pm 0.39$  MPa for the less viscous, especially when taking the production of the samples into account, which was done via injection moulding. Throughout this process pressure is applied, which should increase the wetting, but the viscosity of the matrix plays still a role. Another thing to note is the reduction of the scattering, expressed in terms of standard deviation here, which is considerably smaller for the interfacial shear strengths calculated from the mechanical properties. This is obvious, thinking of the structure of the composites, which were subjected to the mechanical testing. In these composites, many fibres and interfaces are strained, which results in a type of mean anyway. Furthermore, the production is done with injection moulding, where the applied pressure increases the wetting of the fibres again. In single fibre pull out, where just one fibre is strained at a time, the influence of the fibre surface is much bigger, and because we are using natural fibres there has to be some influence thereof.

Table 2. Data from single fibre pull out tests for sisal fibres from different matrices

matrix	C compatibilizer	$\tau_c$	s
	[wt%]	[MPa]	[MPa]
	0	6,21	2,3
	3	9,12	2,78
HD120MO	5	10,13	3,59
	8	10,9	6,36
	25	12,76	5,45
	50	13,51	5,2
	100	12,58	3,36
HC205TF	0	4,58	1,11

The compatibilizer, which was added to the polymer for improving the interfacial shear strength, shows great effectiveness. The improvement is, depending on the concentration, up to 100% increase, which could be found from the data from mechanical testing for both polypropylene types.

For comparing the interfacial shear strength from the composites containing compatibilizer, the first step to do was to recalculate the compatibilizer concentration  $C_c$ , which is the concentration in 100% formulation, to a compatibilizer concentration in the matrix itself,  $C_m$ . Using this concentration, and comparing the interfacial shear strengths with the one from the single fibre pull out tests, one can see a good accordance, as shown in Figure 2. The only data point from mechanical testing, which shows considerably lower interfacial shear strength, is the one acquired from the sample containing 10wt% of sisal fibre in polypropylene HD120MO without compatibilizer. In this case, the low interfacial shear strength in combination with the low fibre content results more in distortion than in reinforcement of the composite, which explains the low value for tensile strength and successive interfacial shear strength.

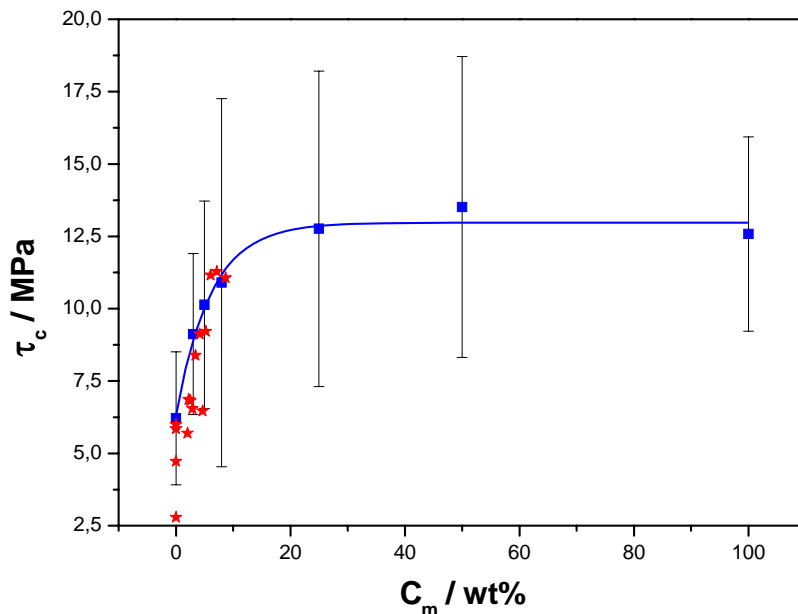


Fig. 2. Interfacial shear strength  $\tau_c$  versus compatibilizer concentration in the matrix  $C_m$  for sisal in polypropylene HD120MO, acquired from single fibre pull out tests (blue) and from the evaluation of tensile strength (red)

#### 4. CONCLUSIONS

As we showed in this work, the interfacial shear strength of sisal fibre reinforced polypropylene composites can be accessed through different pathways. On the one hand, evaluation of the mechanical properties can give reliable data, but suffer from the high effort of extracting the fibres from the composite and the fibre geometry determination. On the other hand, the single fibre pull out test can access the information about the interface, but the influence of sample preparation as well as the influences due to the origin and the surface of the fibres themselves must be considered when interpreting the data.

The polypropylene grade also shows a significant influence on the interfacial shear strength. The less viscous grade wets the fibre better, which results in increased shear strength, as can be seen from the single fibre pull out as well as from the mechanical tests. The addition of a compatibilizer improves the adhesion in a great manner, up to around 100%, depending on the concentration. Further, the results from mechanical testing and the single fibre pull out show very good accordance throughout the whole work.

### ACKNOWLEDGEMENTS

The authors wish to thank Ms. Magdalena Mayer for production and testing of some of the composites as well as Mr. Michael Csombai for doing some work in the single fibre pull out tests.

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